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(19)

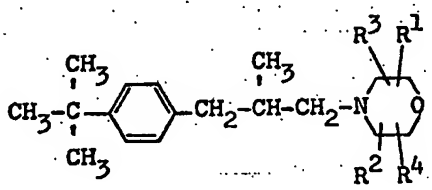
(54) MORPHOLINE DERIVATIVES AND THEIR USE AS FUNGICIDES

(71) We, BASF AKTIENGESELLSCHAFT, a German Joint Stock Company of 6700 Ludwigshafen, Federal Republic of Germany, do hereby declare the invention, for which we pray that a patent may be granted to us, and the method by which it is to be performed, to be particularly described in and by the following Statement:—

The present invention relates to new and valuable morpholine derivatives and their salts and molecular compounds and adducts, having a good fungicidal action, fungicides containing these compounds, and processes for combatting fungi with these compounds.

The use of N - tridecyl - 2,6 - dimethylmorpholine and its salts, molecular compounds and adducts as fungicides has been disclosed (German Patent 1,164,152, German Patent 1,173,722, German Laid-Open Application DOS 2,461,513).

We have now found that morpholine derivatives of the formula

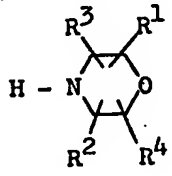


where R^1, R^2, R^3 and R^4 denote hydrogen, methyl or ethyl, and their salts, molecular compounds and adducts have a good fungicidal action.

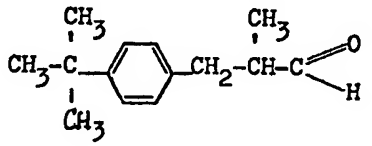
The results tabulated in Examples 3—5 below show that the fungicidal action of compounds according to the invention is superior to that of prior art commercial morpholine derivatives.

Examples of salts are those with inorganic or organic acids, e.g., chlorides, fluorides, bromides, iodides, sulfates, nitrates, phosphates, acetates, propionates and fumarates. Molecular compounds and adducts are formed for instance with acids of surfactants, e.g., dodecylbenzenesulfonic acid.

If the new compounds are 2,6-dimethyl derivatives, they may be isolated as cis and trans isomers. The compounds may be manufactured for instance by reaction of a morpholine of the formula



where R^1 to R^4 have the above meanings, e.g. 2,6 - dimethylmorpholine, with 3 - *p* - tertbutylphenyl - 2 - methylpropanal having the formula



in the presence of a diluent, e.g. formic acid, at from 50° to 110°C.

The preparation of the new compounds is illustrated by the following examples.

EXAMPLE 1

Synthesis of N-(3-*p*-tert-butylphenyl-2-methyl-1-propyl)-cis-2,6-dimethylmorpholine

2,6-Dimethylmorpholine which has been obtained by sulfuric acid-catalyzed cyclization of diisopropanolamine is separated into the cis and trans forms by fractional distillation in a column packed with steel mesh coils. About 75 wt% of the 2,6-dimethylmorpholine is in the cis configuration, the remainder in the trans form. The isomers are separated by fractional distillation in a column having about 40 theoretical plates. The cis form distils as a 99% product at 80° to 81°C and a pressure of 100 mm Hg. The trans form of 2,6-dimethylmorpholine may be obtained under the same conditions at 88° to 89°C/100 mm Hg as a more than 95% product.

575 g of 98% formic acid is placed in a stirred apparatus equipped with reflux condenser, thermometer and dropping funnel. While stirring and cooling, 345 g of 99% 2,6-cis-dimethylmorpholine is then dripped in. The mixture is subsequently slowly heated on a water bath to 70°C. Over a period of 4 hours, 612 g of 3-*p*-tert-butylphenyl-2-methylpropanal is dripped into the reaction mixture, a temperature of approx. 100°C being maintained. Condensation proceeds with marked evolution of CO_2. Upon conclusion of the reaction the mixture is kept for 2 hours at 100°C while stirring.

The excess formic acid is then distilled off under reduced pressure. Substantial separation of the formic acid is achieved under a water pump vacuum at 100°C.

The base is liberated from the formate by dripping in 500 g of 40% aqueous caustic soda solution. The caustic soda solution is advantageously added at from 80° to 100°C to facilitate admixture of the amine phase which forms with the caustic phase. 200 g of toluene is added to depress the viscosity. After separation of the caustic phase, the organic phase is washed twice with water, each time with 250 g.

For further purification, the amine is fractionally distilled at 0.2 mm Hg in a distillation column having 5 trays. In addition to a small amount of first runnings (up to 143°C/0.2 mm Hg; 50 g), there is obtained 865 g of N - (3 - *p* - tert - butylphenyl) - 2 - methyl - 1 - propyl) - 2,6 - cis - dimethylmorpholine, which distils at 0.2 mm Hg between 143° and 146°C. According to gas-chromatographic analysis, the amine is more than 98% pure. With reference to aldehyde, the yield was 84.5%.

To convert the compound to the hydrochloride, 30 g of the pure product is dissolved in 50 g of ethanol which has been saturated with hydrogen chloride at room temperature. After cooling, there is obtained 23 g of the hydrochloride (m.p. 220°C) in very pure form.

EXAMPLE 2

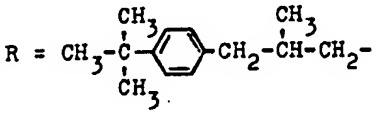
Synthesis of N-(3-*p*-tert-butylphenyl-2-methyl-1-propyl)-2,6-trans-dimethylmorpholine

While cooling with ice, 29 g of 2,6-trans-dimethylmorpholine is introduced into 70 g of 98% formic acid, while stirring, 41 g of 3 - *p* - tert - butylphenyl - 2 - methylpropanal is then added. The reaction mixture is heated for 6 hours at 100°C. Upon commencement of the reaction, very strong CO_2 evolution takes place which drops noticeably after about an hour. Further working up is effected as disclosed in more detail in Example 1.

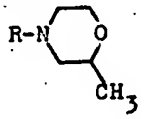
N - (3 - *p* - tert - butylphenyl - 2 - methyl - 1 - propyl) - 2,6 - trans - dimethylmorpholine distils at 168°—169°C (5 mm Hg). The yield is 52 g (86% of theory, with reference to aldehyde). 11 g of the amine is dissolved in 20 g of ethyl acetate which has been saturated with anhydrous hydrogen chloride. The hydrochloride crystallizes out upon cooling; m.p.: 165°C.

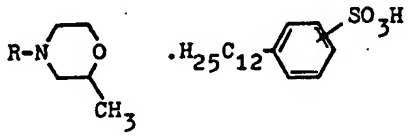
The other compounds according to the invention may be obtained analogously.

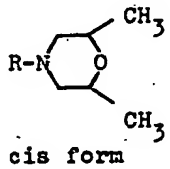
Examples of compounds according to the invention are given below:



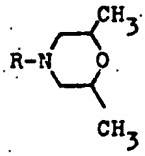
Compound no.

1  b.p.: 123°C/0.01 mm

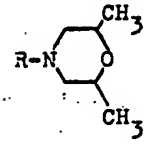
2  m.p.: 108°C

3  b.p.: 143-146°C/0.2 mm

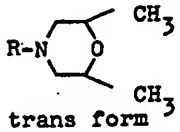
cis form

4  b.p.: 168-169°C/5 mm

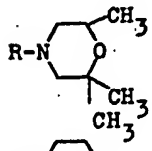
trans form

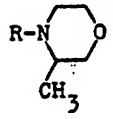
5  .HCl m.p.: 220°C

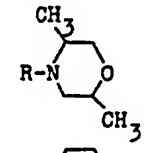
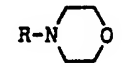
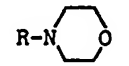
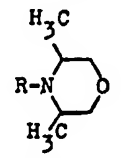
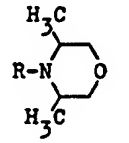
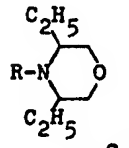
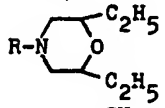
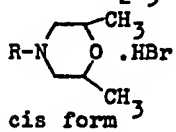
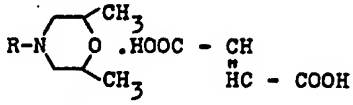
cis form

6  .HCl m.p.: 165°C

trans form

10  b.p.: 128°C/0.02 mm

11  b.p.: 133°C/0.05 mm

12		b.p.: 125°C/0.02 mm
13		b.p.: 170°C/5.0 mm
14		m.p.: 206°C
15		b.p.: 145°C/0.3 mm
16		m.p.: 172°C
17		b.p.: 148°C/0.3 mm
18		b.p.: 146-148°C/0.3 mm
19	 cis form	m.p.: 228°C
20	 cis form	m.p.: 174°C

The morpholine derivatives according to the invention and the fungicides containing them are particularly suitable for combatting plant diseases such as *Erisiphe graminis* in cereals, *Erysiphe cichoriacearum* in Cucurbitaceae, *Podosphaera leucotricha* in apples, *Uncinula necator* in grapes, *Erysiphe polygoni* in beans, *Sphaerotheca pannosa* in roses, *Microsphaera querci* in oaks, *Botrytis cinerea* in strawberries and grapes, *Mycosphaerella musicola* in bananas, Puccinia species (rusts) in cereals, *Uromyces appendiculatus* and *U. phaseoli* in beans, *Hemileia vastatrix* in coffee, and *Rhizoctonia solani*. Therefore, they may be applied, for example, to cereal plants (e.g. wheat and barley for protection against rust and/or mildew), bean plants, apple trees, grape vines, cucurbitaceae plants, rose trees, oak trees, strawberry plants, banana trees and coffee plants. They have a systemic action; they are not only taken up by the roots but also absorbed by the leaves, and are translocated in the plant tissue. They may be used to treat the plants after fungus attack has begun.

Application is effected for instance by watering, spraying, dusting, or disinfecting the plants or treating the seed with the morpholine derivatives.

When the morpholine derivatives are used to protect plants against fungus infection, the application rates are from 0.025 to 5 kg of active ingredient per hectare. For the surface protection of trees or fruit, the morpholine derivatives may also be used in combination with plastics dispersions in amounts of from 0.25 to 5%, based on the weight of the dispersion. In general, the fungicidal compositions according to the invention contain from 0.1 to 95, preferably from 0.5 to 90, wt% of the morpholine derivative.

The morpholine derivatives may also be mixed with other, prior art, fungicides. In many instances, the spectrum of fungicidal action is increased; with a number of fungicidal compositions in the weight ratio range of 1:10 to 10:1 synergistic effects also occur, i.e., the fungicidal action of the combination product is greater than the effect of the individual components added together. Examples of fungicides which may be combined with the morpholine derivatives of the invention are dithiocarbamates and derivatives thereof, e.g.,

zinc dimethyldithiocarbamate
manganese ethylenebisdithiocarbamate
zinc ethylenebisdithiocarbamate
tetramethylthiuram disulfide
ammonia complex of zinc-(N,N'-ethylene-bisdithiocarbamate) and N,N'-polyethylene-bis-(thiocarbamoyl)-disulfide
ammonia complex of zinc-(N,N'-propylene-bisdithiocarbamate) and N,N'-polypropylene-bis-(thiocarbamoyl)-disulfide

heterocyclic structures, such as

N-trichloromethylthiotetrahydrophthalimide
N-trichloromethylthiophthalimide
N-(1,1,2,2-tetrachloroethylthio)-tetrahydrophthalimide
methyl 1-(butylcarbamoyl)-2-benzimidazole carbamate
2-methoxycarbonylaminobenzimidazole
2,3-dihydro-5-carboxanilido-6-methyl-1,4-oxathiin-4,4-dioxide
2,3-dihydro-5-carboxanilido-6-methyl-1,4-oxathiin
5-butyl-2-dimethylamino-4-hydroxy-6-methylpyrimidine
1,2-bis-(3-ethoxycarbonyl-2-thioureido)-benzene
1,2-bis-(3-methoxycarbonyl)-2-thioureido)-benzene

and various fungicides, such as

dodecylguanidine acetate
N-dichlorofluoromethylthio-N',N'-dimethyl-N-phenylsulfuric acid diamide
2,5-dimethylfuran-3-carboxylic acid anilide
2,5-dimethylfuran-3-carboxylic acid cyclohexyl amide
2-iodobenzoic acid anilide
2-bromobenzoic acid anilide
3-nitroisophthalic acid diisopropyl ester
1-(1,2,4-triazolyl-1')-[1-(4'-chlorophenoxy)]-3,3-dimethylbutan-2-one
1-(1-imidazolyl)-2-allyloxy-2-(2,4-dichlorophenyl)-ethane
piperazine-1,4-diyl-bis-1-(2,2,2-trichloroethyl)-formamide
2,4,5,6-tetrachloroisophthalonitrile
1,2-dimethyl-3,5-diphenylpyrazoliniummethylsulfate.

Application may be effected for instance in the form of directly sprayable solutions, powders, suspensions, dispersions, emulsions, oil dispersions, pastes, dusts, broadcasting agents, or granules by spraying, atomizing, dusting, broadcasting or watering. The forms of application depend entirely on the purpose for which the morpholine derivatives are being used; in any case they should ensure a fine distribution of the morpholine derivative.

For the preparation of solutions, emulsions, pastes and oil dispersions to be sprayed direct, mineral oil fractions of medium to high boiling point, such as kerosene or diesel oil, further coal-tar oils, and oils of vegetable or animal origin, aliphatic, cyclic and aromatic hydrocarbons such as benzene, toluene, xylene, paraffin, tetrahydronaphthalene and alkylated naphthalenes, and hydrocarbon derivatives such as methanol, ethanol, propanol, butanol, chloroform, carbon tetrachloride, cyclohexanol, cyclohexanone, chlorobenzene and isophorone, as

well as strongly polar solvents such as dimethylformamide, dimethyl sulfoxide, N-methylpyrrolidone and water, are suitable.

Aqueous formulations may be prepared from emulsion concentrates, pastes, oil dispersions or wettable powders by adding water. To prepare emulsions, pastes and oil dispersions the ingredients as such or dissolved in an oil or solvent may be homogenized in water by means of wetting or dispersing agents, adherents or emulsifiers. Concentrates which are suitable for dilution with water may be prepared from the morpholine derivative wetting agent, adherent, emulsifying or dispersing agent and possibly solvent or oil.

Examples of surfactants are: alkali metal, alkaline earth metal and ammonium salts of ligninsulfonic acid, naphthalenesulfonic acids, phenolsulfonic acids, alkylaryl sulfonates, alkyl sulfates, and alkyl sulfonates, alkali metal and alkaline earth metal salts of dibutylnaphthalenesulfonic acid, lauryl ether sulfate, fatty alcohol sulfates, alkali metal and alkaline earth metal salts of fatty acids, salts of sulfated hexadecanols, heptadecanols, and octadecanols, salts of sulfated fatty alcohol glycol ethers, condensation products of sulfonated naphthalene and naphthalene derivatives with formaldehyde, condensation products of naphthalene or naphthalenesulfonic acids with phenol and formaldehyde, polyoxyethylene octylphenol ethers, ethoxylated isooctylphenol, ethoxylated octylphenol and ethoxylated nonylphenol, alkylphenol polyglycol ethers, tributylphenol polyglycol ethers, alkylaryl polyester alcohols, isotridecyl alcohols, fatty alcohol ethylene oxide condensates, ethoxylated castor oil, polyoxyethylene alkyl ethers, ethoxylated polyoxypropylene, lauryl alcohol polyglycol ether acetal, sorbitol esters, lignin, sulfite waste liquors and methyl cellulose.

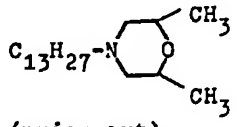
Powders, dusts and broadcasting agents may be prepared by mixing or grinding the morpholine derivatives with a solid carrier.

Granules, e.g., coated, impregnated or homogeneous granules, may be prepared by bonding the morpholine derivatives to solid carriers. Examples of solid carriers are mineral earths such as silicic acid, silica gels, silicates, talc, kaolin, Attaclay, limestone, lime, chalk, bole, loess, clay, dolomite, diatomaceous earth, calcium sulfate, magnesium sulfate, magnesium oxide, ground plastics, fertilizers such as ammonium sulfate, ammonium phosphate, ammonium nitrate, and ureas, and vegetable products such as grain flours, bark meal, wood meal, and nutshell meal, cellulosic powders, etc.

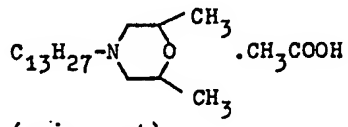
There may be added to the compositions or individual active ingredients oils of various types, herbicides, fungicides, nematocides, insecticides, bactericides, trace elements, fertilizers, antifoams (e.g., silicones), growth regulators, antidotes and other herbicidally effective compounds.

The following experiments, the following prior art compounds were used for comparison purposes:

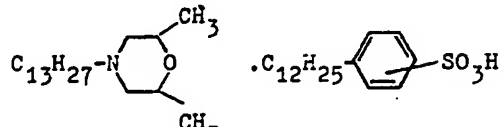
Compound no.

7 

(prior art)

8 

(prior art)

9 

(prior art)

EXAMPLE 3

Leaves of wheat seedlings of the "Jubilar" variety grown in pots are treated with aqueous emulsions consisting of 80% (wt%) of active ingredient and 20% emulsifier and dusted, after the sprayed-on layer has dried, with spores of wheat mildew (*Erysiphe graminis var. tritici*). The plants are then placed in a greenhouse at from 20° to 22°C and 75 to 80% relative humidity. The extent the fungus has spread is determined after 10 days.

Active Ingredient	Leaf Attack After Spraying With Liquors Containing Active Ingredients in Amounts of 0.006%	0.012%	0.025%	0.05%
1	1—2	1	0—1	0
2	1	0—1	0	0
3	0	0	0	0
4	1	0	0	0
5	0	0	0	0
6	1	0	0	0
19	0	0	0	0
20	0	0	0	0
7 } prior art	3—4	3	2	1
8 } prior art	4	4	2	1
9 } prior art	2	1	1	0
Control (untreated)	4			

0=no attack, graduated down to 5=surface of leaves completely covered by fungus.

EXAMPLE 4

Leaves of barley seedlings of the "Firlbecks Union" variety grown in pots are treated as described in Example 3, and dusted with spores of barley mildew (*Erysiphe graminis var. hordei*).

Active Ingredient	Leaf Attack After Spraying With Liquors Containing Active Ingredient in Amounts of 0.006%	0.012%	0.025%
1	0	0	0
2	0	0	0
3	0	0	0
4	0	0	0
5	0	0	0
6	0	0	0
19	0	0	0
20	0	0	0
7 } prior art	2	1	0—1
8 } prior art	3	1	1
Control (untreated)	4		

0=no attack, graduated down to 5=surface of leaves completely covered by fungus.

EXAMPLE 5

Leaves of wheat plants grown in pots are artificially infected with spores of leaf rust (*Puccinia recondita*) and placed for 48 hours in a steam-saturated chamber kept at 20° to 25°C. The plants are then sprayed with aqueous liquors containing a mixture, dissolved or emulsified in water, of 80% of the active ingredient and 20% of sodium lignin sulfonate, and placed in a greenhouse at from 20° to 22°C and 75 to 80% relative humidity. The extent the fungus has spread is assessed after 10 days.

Active Ingredient	Leaf Attack After Spraying With Liquor Containing Active Ingredient in Amounts of 0.025	0.05	0.1
1	2—3	1—2	0
3	0	0	0
4	0	0	0
5	0	0	0
6	0	0	0
19	0	0	0
20	0	0	0
7 } prior art	4	3	2
8 } prior art	4	4	3
9 } prior art	4	3	3
Control (untreated)	4		

0=no damage, graduated down to 5=surface of leaves completely covered by fungus.

EXAMPLE 6

90 parts by weight of compound 3 is mixed with 10 parts by weight of N-methyl-α-pyrrolidone. A mixture is obtained which is suitable for application in the form of very fine drops.

EXAMPLE 7

20 parts by weight of compound 4 is dissolved in a mixture consisting of 80 parts by weight of xylene, 10 parts by weight of the adduct of 8 to 10 moles of ethylene oxide to 1 mole of oleic acid N-monoethanolamide, 5 parts by weight of the calcium salt of dodecylbenzenesulfonic acid, and 5 parts by weight of the adduct of 40 moles of ethylene oxide to 1 mole of castor oil. By pouring the solution into 100,000 parts by weight of water and uniformly distributing it therein, an aqueous dispersion is obtained containing 0.02% by weight of the active ingredient.

EXAMPLE 8

20 parts by weight of compound 3 is dissolved in a mixture consisting of 40 parts by weight of cyclohexanone, 30 parts by weight of isobutanol, 20 parts by weight of the adduct of 7 moles of ethylene oxide to 1 mole of isooctylphenol, and 10 parts by weight of the adduct of 40 moles of ethylene oxide to 1 mole of castor oil. By pouring the solution into 100,000 parts by weight of water and uniformly distributing it therein, an aqueous dispersion is obtained containing 0.02% by weight of the active ingredient.

EXAMPLE 9

20 parts by weight of compound 4 is dissolved in a mixture consisting of 25 parts by weight of cyclohexanol, 65 parts by weight of a mineral oil fraction having a boiling point between 210° and 280°C, and 10 parts by weight of the adduct of 40 moles of ethylene oxide to 1 mole of castor oil. By pouring the solution into 100,000 parts by weight of water and uniformly distributing it therein, an aqueous dispersion is obtained containing 0.02% by weight of the active ingredient.

EXAMPLE 10

20 parts by weight of compound 5 is well mixed with 3 parts by weight of the sodium salt of diisobutylnaphthalene-α-sulfonic acid, 17 parts by weight of the sodium salt of a ligninsulfonic acid obtained from a sulfite waste liquor, and 60 parts by weight of powdered silica gel, and triturated in a hammer mill. By uniformly distributing the mixture in 20,000 parts by weight of water, a spray liquid is obtained containing 0.1% by weight of the active ingredient.

EXAMPLE 11

3 parts by weight of compound 3 is intimately mixed with 97 parts by weight of particulate kaolin. A dust is obtained containing 3% by weight of the active ingredient.

EXAMPLE 12

30 parts by weight of compound 4 is intimately mixed with a mixture consisting of 92 parts by weight of powdered silica gel and 8 parts by weight of paraffin oil which has been sprayed onto the surface of this silica gel. A formulation of the active ingredient is obtained having good adherence.

EXAMPLE 13

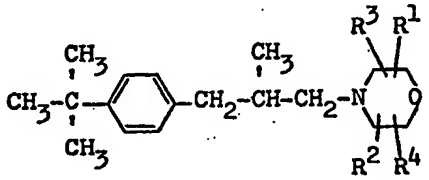
40 parts by weight of compound 3 is intimately mixed with 10 parts of the sodium salt of a phenolsulfonic acid-urea-formaldehyde condensate, 2 parts of silica gel and 48 parts of water to give a stable aqueouos dispersion. Dilution in 100,000 parts by weight of water gives an aqueous dispersion containing 0.04 wt% of active ingredient.

EXAMPLE 14

20 parts of compound 4 is intimately mixed with 2 parts of the calcium salt of dodecylbenzenesulfonic acid, 8 parts of a fatty alcohol polyglycol ether, 2 parts of the sodium salt of a phenolsulfonic acid-urea-formaldehyde condensate and 68 parts of a paraffinic mineral oil. A stable oily dispersion is obtained.

WHAT WE CLAIM IS:—

1. A morpholine derivative of the formula



where R^1, R^2, R^3 and R^4 each independently denote hydrogen, methyl or ethyl, or a salt, molecular compound or adduct thereof.

2. N - (3 - *p* - tert - butylphenyl - 2 - methyl - 1 - propyl) - 2,6 - dimethylmorpholine.

3. N - (3 - *p* - tert - butylphenyl - 2 - methyl - 1 - propyl) - 2 - methylmorpholine.

4. N - (3 - *p* - tert - butylphenyl - 2 - methyl - 1 - propyl) - 3 - methylmorpholine.

5. N - (3 - *p* - tert - butylphenyl - 2 - methyl - 1 - propyl) - morpholine.

6. N - (3 - *p* - tert - butylphenyl - 2 - methyl - 1 - propyl) - 2,2,6 - trimethylmorpholine.

7. N - (3 - *p* - tert - butylphenyl - 2 - methyl - 1 - propyl) - 2,5 - dimethylmorpholine.

8. N - (3 - *p* - tert - butylphenyl - 2 - methyl - 1 - propyl) - 3,5 - dimethylmorpholine.

9. N - (3 - *p* - tert - butylphenyl - 2 - methyl - 1 - propyl) - 2,6 - diethylmorpholine.

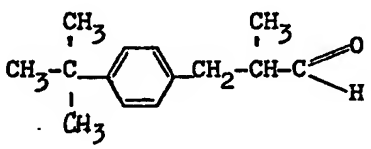
10. N - (3 - *p* - tert - butylphenyl - 2 - methyl - 1 - propyl) - 3,5 - diethylmorpholine.

11. N - (3 - *p* - tert - butylphenyl - 2 - methyl - 1 - propyl) - cis - 2,6 - dimethylmorpholine.

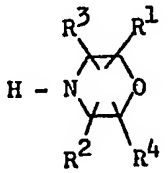
12. A morpholine derivative as claimed in any of claims 1 to 11 in the form of a molecular compound or adduct with an acid of a surfactant or in the form of a salt with an inorganic or organic acid.

13. A morpholine derivative as claimed in claim 12 in the form of its chloride, fluoride, bromide, iodide, sulfate, nitrate, phosphate, acetate, propionate or fumarate salt or its dodecylbenzenesulfonic acid adduct or molecular compound.

14. A process for preparing a morpholine derivative as claimed in claim 1 wherein 3 - *p* - tert - butylphenyl - 2 - methylpropanal of the formula



is reacted with a morpholine of the formula



where R^1, R^2, R^3 and R^4 have the meanings given in claim 1, in the presence of a diluent at a temperature of from 50° to 110°C.

15. A morpholine derivative as claimed in claim 1 when prepared by a process as claimed in claim 14, or a salt, molecular compound or adduct thereof.

16. The use of a compound as claimed in any of claims 1 to 13 or 15 as a fungicide (other than for application to human beings).

17. A fungicidal composition comprising a solid or liquid carrier and a compound as claimed in any of claims 1 to 13 or 15.

18. A fungicidal composition as claimed in claim 17 in the form of a solution, emulsion, paste or oil dispersion in an organic liquid to be sprayed direct or diluted with water for spraying.

19. A fungicidal composition as claimed in claim 17 as an aqueous formulation.

20. A fungicidal composition comprising a compound as claimed in any of claims 1 to 13 or 15 and a wetting or dispersing agent, adherent or emulsifier.

21. A fungicidal composition as claimed in claim 17 in the form of a dust, wettable powder, broadcastable composition or granules.

22. A fungicidal composition as claimed in any of claims 17 to 21 which contains a further fungicide.

23. A fungicide composition comprising a compound as claimed in any of claims 1 to 13 or 15 and a further fungicidal compound listed herein.

24. A process for preparing a fungicidal composition, wherein a solid or liquid carrier is mixed with a compound as claimed in any of claims 1 to 13 or 15.

25. A process for combatting fungi, wherein the locus to be protected against fungus attack (which locus is not a human being) is treated with a compound as claimed in any of claims 1 to 13 or 15.

26. A process for combatting fungal attack in plants wherein the plants, or their seeds, are treated with a compound as claimed in any of claims 1 to 13 or 15.

27. A process as claimed in claim 26 wherein the plants are treated after fungus attack.

28. A process as claimed in claim 26 or 27 wherein the plants are cereal plants, bean plants, apple trees, grape vines, cucurbitaceae plants, rose trees, oak trees, strawberry plants, banana trees, or coffee plants.

29. A process as claimed in claim 28 wherein wheat or barley plants are treated for protection against rust and/or mildew.

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